WHAT IS CLAIMED IS:

1. A method of forming a multilayer thin film heterostructure comprising:

applying a solution including a first water-soluble polymer selected from the group consisting of polyanionic species, polycationic species and uncharged polymer species onto a spinning substrate to form a first coating layer on said substrate;

drying said first coating layer on said substrate;

applying a solution including a second water-soluble polymer selected from the group consisting of polyanionic species, polycationic species and uncharged polymer species onto said substrate having said first coating layer thereon to form a second coating layer on said first coating layer, said second water-soluble polymer characterized as a different material than said first water-soluble polymer; and,

drying said second coating layer on said first coating layer, so that a bilayer is built up upon said substrate.

2. The method of claim 1 wherein said process further includes repeating one or more additional applying and drying sequence with a water-soluble polymer selected from the group consisting of polyanionic species, polycationic species and uncharged polymer species, so that a predetermined plurality of layers are built up upon said substrate.

3. The method of claim 1 wherein said bilayer has a polycationic layer/polyanionic layer structure.

4. The method of claim 2 wherein said plurality of layers includes multiple bilayers having a polycationic layer/polyanionic layer structure.

5. The method of claim 2 wherein said plurality of layers includes multiple bilayers having a polyanionic layer/polycationic layer structure.

6. The method of claim 2 wherein said plurality of layers includes multiple trilayers having a polycationic layer/polyanionic layer/polyanionic layer structure.

7. The method of claim 2 wherein said plurality of layers includes multiple trilayers having a polycationic layer/polyanionic layer/uncharged polymer layer structure

8. The method of claim 1 wherein said drying steps comprise subjecting said coated substrate to a vacuum for sufficient time to effect drying of said coating layers.

9. The method of claim 1 wherein said drying steps comprise heating said coated substrate at a predetermined temperature for sufficient time to effect drying of said coating layers.

10. The method of claim 1 wherein said polycationic species are selected from the group consisting of polyethylenimine, poly(diallyldimethyl ammonium chloride), poly(allylamine hydrochloride), and poly(propylenimine) dendrimers.

11. The method of claim 1 wherein said polyanionic species are selected from the group consisting of poly[1-[4-(3-carboxy-4-hydroxy-phenylazo)benzene sulfonamido]-1,2-ethanediyl, sodium salt], poly(acrylic acid), poly(styrenesulfonate), poly(4-[4-({4-[3-amino-2-(4-hydroxy-phenyl)-propylcarbamoyl]-5-oxo-pentyl}-methyl-amino)-phenylazo]-benzenesulfonic acid).

12. The method of claim 1 wherein at least one solution further includes a surfactant and a resultant coating layer from said solution including said surfactant further includes said surfactant.

13. The method of claim 1 wherein at least one solution further includes a dye molecule and a resultant coating layer from said solution including said dye molecule further includes said dye molecule.

14. The method of claim 1 wherein said uncharged polymer species are selected from the group consisting of poly(vinylpyrrolidinone), polysaccharides, and biopolymers.

15. The method of claim 6 wherein trilayer thicknesses in said polycationic layer/polyanionic layer/polyanionic layer structure are about equal.

16. The method of claim 1 wherein at least one water-soluble polymer includes a chromophore.

17. The method of claim 16 wherein said multilayer thin film heterostructure is a non-linear optical structure.